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Surface plasmon polaritons in dielectric/metal/dielectric structures: metal layer thickness influence

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A model is proposed and studied that makes it possible to explain experimental data on a metal layer thickness influence on the spectrum of SPPs waves in heterogeneous dielectric/metal/dielectric structures.

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1. Introduction

Surface plasmon polaritons (SPPs) are collective excitations of electrons, propagating at the interface between a metal and a dielectric [1, 2]. They are used to manipulate electromagnetic energy at the subwavelength scales, which necessitates the study of their characteristics.

A considerable amount of research on the study of SPPs spectrum in heterogeneous dielectric/metal/dielectric structures has been published for today (see bibliography in [1, 3]), in which a metal layer is mainly considered as 2D metal or metallic graphene using the corresponding characteristic expressions for the dielectric function $\varepsilon(\boldsymbol{q},\omega)$ ($\boldsymbol{q}=(k_x,k_y)$ is 2D vector, ω is frequency) of a metal.

In the case when a metal layer is a 3D structure, the Drude model is widely used to describe SPPs [1, 4] in which the dielectric permittivity of a metal is expressed by the formula:

$$\begin{gathered}\varepsilon(\boldsymbol{q},z,z',\omega)=\varepsilon^D(\omega)\,\delta(z-z'),\\ \varepsilon^D(\omega)=1-\frac{\omega_p^2}{\omega^2+i\gamma\omega}.\end{gathered}\tag{1}$$

Unfortunately, this approach to the description of SPPs does not allow taking into account the influence of the thickness of a metal layer and size effects on their properties.

Recently, in the paper [5] there has been presented the results of experimental studies on the influence of a thickness of a metal film on the spectrum $\hbar\omega$ ($\hbar$ is the reduced Planck constant [6]) of SPPs, where it has been demonstrated that such a dependence is significant in the area of small ($\sim 1-5\,\text{nm}$) thicknesses. In this paper, a mathematical model for $\varepsilon(\boldsymbol{q},\omega,z,z')$ is proposed and studied and it is shown that taking into account the thickness of a metal layer can be described by such a model; and the obtained results qualitatively coincide with the experimental results.

2. Problem formulation

Consider a heterogeneous structure (whose geometry is depicted in Fig. 1) formed by two non-conducting media with dielectric permittivities ε_1 and ε_3 between which a metal nanofilm with thickness L is sandwiched. We assume that dielectric permittivities ε_1 and ε_3 are functions of the time variable, i.e.

$$\varepsilon_1=\varepsilon_1(t),\quad \varepsilon_3=\varepsilon_3(t).\tag{2}$$

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Dielectric permittivity of metal is $\varepsilon_2(\boldsymbol{r},\boldsymbol{r}',t)$ and for the geometry of a dielectric/metal/dielectric heterostructure has the form

$$\varepsilon_2=\varepsilon_2(\boldsymbol{r}_{||}-\boldsymbol{r}'_{||},z,z',t),\quad \boldsymbol{r}_{||}=(x,y). \tag{3}$$

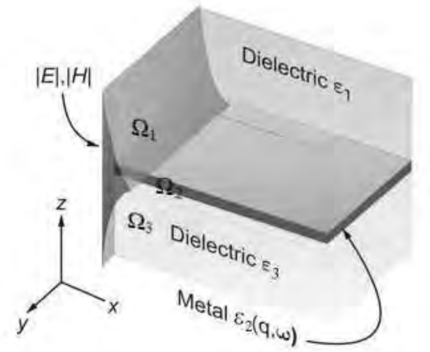

Fig. 1. Schematic representation of dielectric/metal/dielectric structure.

Let us consider a problem of describing the propagation of electromagnetic waves, which are localized at the interface between a dielectric ($z \geqslant L$) and a metal ($0<z<L$). These waves are called surface plasmons [1]. A mathematical model describing the propagation of surface plasmons is based on the Maxwell's equations system [1, 2]:

$$\begin{gathered}\nabla\cdot\boldsymbol{D}=\rho,\quad \nabla\cdot\boldsymbol{B}=0,\\ \nabla\times\boldsymbol{E}=\rho,\quad \nabla\times\boldsymbol{H}=\boldsymbol{J}+\frac{\partial\boldsymbol{D}}{\partial t},\end{gathered} \tag{4}$$

where $\boldsymbol{D}$ is electric flux density, $\boldsymbol{B}$ is magnetic flux density, $\boldsymbol{E}$ is electric field strength, $\boldsymbol{H}$ is magnetic field strength, ρ is electric charge density and $\boldsymbol{J}$ is electric current density. We assume that external charges ρ in the area of contact between dielectrics and metal are absent, namely $\nabla\cdot\boldsymbol{D}=0$. Here "$\cdot$" is the dot product, "$\times$" is the cross product.

We assume that interconnection between the vectors $\boldsymbol{E}$ and $\boldsymbol{D}$ [2], namely

$$\boldsymbol{D}(\boldsymbol{r}_{||},z,t)=\iiint d\boldsymbol{r}'_{||}\,dz'\,dt'\,\varepsilon_i(\boldsymbol{r}_{||}-\boldsymbol{r}'_{||},z,z',t-t')\,\boldsymbol{E}(\boldsymbol{r}'_{||},z',t'),\quad i=1,2,3. \tag{5}$$

Let us write the system of Maxwell's equations (4) in Fourier variables. We will define the Fourier transform with respect to time as

$$f(t)=\frac{1}{2\pi}\int_{-\infty}^{\infty}\tilde{f}(\omega)\,e^{i\omega t}d\omega,\quad \tilde{f}(\omega)=\int_{-\infty}^{\infty}f(t)\,e^{-i\omega t}dt. \tag{6}$$

And $\varepsilon_i(\boldsymbol{r}_{||}-\boldsymbol{r}'_{||},z,z',t-t')$ is expressed by the following equation:

$$\varepsilon_i(\boldsymbol{r}_{||}-\boldsymbol{r}'_{||},z,z',t-t')=\frac{\Omega}{(2\pi)^3}\int_{-\infty}^{\infty}d\omega\int_{\Omega}d\boldsymbol{q}\,\varepsilon_i(\boldsymbol{q},z,z',\omega)\,e^{-i(\boldsymbol{q},\boldsymbol{r}_{||}-\boldsymbol{r}'_{||})-i\omega(t-t')}, \tag{7}$$

where Ω is the domain of the 2D vector $\boldsymbol{q}=(k_x,k_y)$.

We will assume that

$$\begin{aligned}\varepsilon_1(\boldsymbol{r}_{||}-\boldsymbol{r}'_{||},z,z',t-t')&=\varepsilon_1(t-t')\,\delta(\boldsymbol{r}_{||}-\boldsymbol{r}'_{||})\,\delta(z-z'),\\ \varepsilon_2(\boldsymbol{r}_{||}-\boldsymbol{r}'_{||},z,z',t-t')&=\varepsilon_2(\boldsymbol{r}_{||}-\boldsymbol{r}'_{||},z,z,t-t')\,\delta(z-z'),\\ \varepsilon_3(\boldsymbol{r}_{||}-\boldsymbol{r}'_{||},z,z',t-t')&=\varepsilon_3(t-t')\,\delta(\boldsymbol{r}_{||}-\boldsymbol{r}'_{||})\,\delta(z-z'),\end{aligned} \tag{8}$$

where $\delta(z-z')$ is the Dirac delta function [6].

The polarization of the waves corresponds to the transverse magnetic (TM) mode for the vectors $\boldsymbol{E}$ and $\boldsymbol{H}$, that is

$$\boldsymbol{E}=(E_x,0,E_y),\quad \boldsymbol{H}=(0,H_y,0). \tag{9}$$

Consequently [1], the magnetic field propagates along the axis OX and is homogeneous along the axis OY

$$\boldsymbol{H}(\boldsymbol{r},\omega)=\boldsymbol{H}(z,\omega)\,e^{ik_x x}, \tag{10}$$

k_x is a wave vector in the direction of propagation.

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For $\boldsymbol{H}$ we obtain a system of wave equations for all the domains of the heterogeneous structure [1]:

$$\frac{\partial^2 H_y(z,\omega)}{\partial z^2} + \left(k_0^2\,\varepsilon_1(\omega) - k_x^2\right)H_y(z,\omega) = 0, \tag{11}$$

$$\frac{\partial^2 H_y(z,\omega)}{\partial z^2} + \left(k_0^2\,\varepsilon_2(\boldsymbol{q},z,z,\omega) - k_x^2\right)H_y(z,\omega) = 0, \tag{12}$$

$$\frac{\partial^2 H_y(z,\omega)}{\partial z^2} + \left(k_0^2\,\varepsilon_3(\omega) - k_x^2\right)H_y(z,\omega) = 0, \tag{13}$$

where $k_0 = \omega/c$. In order to solve the system (11)–(13), we need to find out expressions for dielectric permittivities $\varepsilon_1(\omega)$, $\varepsilon_2(\boldsymbol{q},z,z,\omega)$, and $\varepsilon_3(\omega)$.

3. Model of dielectric permittivity of a metal layer

Here and subsequently, we will use a high-frequency approximation for dielectric layers, this implies that in (8) the first and the last expressions can be rewritten as follows

$$\begin{aligned} \varepsilon_1(\omega) &= \varepsilon_1(\infty) = \varepsilon_1 = \text{const}, \\ \varepsilon_3(\omega) &= \varepsilon_3(-\infty) = \varepsilon_3 = \text{const}. \end{aligned} \tag{14}$$

As a model for the dielectric function $\varepsilon_2(\boldsymbol{q},z,z',\omega)$ of the metal layer, we will use the diagonal component of the dielectric permittivity tensor of a metal film which is obtained in [7],

$$\varepsilon(\boldsymbol{r},\boldsymbol{r}',\omega) = \left(1 - \frac{\omega_p^2}{n_e\omega^2}\sum_n f_n|\psi_n(\boldsymbol{r}')|^2\right)\delta(\boldsymbol{r}-\boldsymbol{r}'). \tag{15}$$

Here $\omega_p = \sqrt{4\pi n_e e^2/m_e}$ is the plasma frequency [1, 4], n_e is an electron density in a metal, $f_n = \Theta(\varepsilon_n - \varepsilon_F)$ is the Fermi–Dirac function [7, 8], $\Theta(x)$ is the Heaviside step function [8], ε_F is the Fermi energy [4, 8], $\boldsymbol{r} = (\boldsymbol{r}_{||}, z)$.

The function

$$\psi_n(x,y,z) = \sqrt{\frac{2}{S}}\,e^{i(\boldsymbol{q}\cdot\boldsymbol{r}_{||})}\phi_n(z) \tag{16}$$

is a wave function [6] of an electron in the metal layer and $\phi_n(z)$ is the solution of equation

$$-\frac{\hbar^2}{2m}\frac{d^2}{dz^2}\phi_n(z) + U(z)\phi_n(z) = W\phi_n(z), \tag{17}$$

which is the Schrödinger equation [3] that describes behaviour of an electron in a metal film [6, 9, 10]. Potential $U(z)$ that simulates surfaces bounding the film has the form

$$U(z) = \begin{cases} U_1 & \text{if} \quad z < 0, \\ 0 & \text{if} \quad 0 < z < L, \\ U_2 & \text{if} \quad z > L, \end{cases} \tag{18}$$

where L is the film thickness. The solutions of the equation (17) that satisfies the conditions $\phi(z \to \pm\infty) \to 0$ can be presented as follows

$$\phi_n(z) = \begin{cases} A\,e^{\chi_1 z}, \quad \chi_1 = \sqrt{\frac{2m}{\hbar^2}(U_1 - W)} & \text{if} \quad z < 0, \\ C_1\,e^{ikz} + C_2\,e^{-ikz}, \quad k = \sqrt{\frac{2m}{\hbar^2}W} & \text{if} \quad 0 < z < L, \\ B\,e^{-\chi_1 z}, \quad \chi_2 = \sqrt{\frac{2m}{\hbar^2}(U_2 - W)} & \text{if} \quad z > L. \end{cases} \tag{19}$$

Constants A, C_1, C_2, and B we will determine using continuity conditions for $\phi_n(z)$ and $\frac{d\phi_n(z)}{dz}$ on the boundaries $z = 0$ and $z = L$ and a normalization condition

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$$\int_{-\infty}^{\infty} |\phi_n(z)|^2 = 1, \tag{20}$$

which, actually, yields the condition $\phi(z \to \pm\infty) \to 0$. Hence, the expressions for constants have the form

$$C_1 = A\left(\frac{1}{2} - \frac{i\chi_1}{2k}\right), \quad C_2 = A\left(\frac{1}{2} + \frac{i\chi_1}{2k}\right). \tag{21}$$

$$B = A\left(\cos(kL) + \frac{\chi_1}{k}\sin(kL)\right)e^{\chi_2 L}, \tag{22}$$

$$|A|^2 = \left(\frac{1}{2\chi_1} + \frac{1}{2\chi_2}\left(\cos(kL) + \frac{\chi_1}{k}\sin(kL)\right)^2 + \frac{L}{2}\left(\frac{\chi_1^2}{k^2} + 1\right) + \frac{\chi_1}{2k^2}\Big(1 - \cos(2kL)\Big) + \frac{1}{4}\left(\frac{1}{k} - \frac{\chi_1^2}{k^3}\right)\sin(2kL)\right)^{-1}. \tag{23}$$

In order to find k, we need to solve the following equation [3, 9, 10]

$$kL = \pi n - \left(\arcsin\frac{k\hbar}{\sqrt{2mU_1}} + \arcsin\frac{k\hbar}{\sqrt{2mU_2}}\right), \tag{24}$$

the roots of (24) will determine a value of

$$W_n = \frac{\hbar^2 k_n^2}{2m} \tag{25}$$

which is discrete.

The maximum number of energy levels $n_{\max}$ we determine from the condition

$$n_{\max} = \left[\frac{1}{\pi}\left(L\min(S_1,S_2) + \arcsin\frac{\min(S_1,S_2)}{S_1} + \arcsin\frac{\min(S_1,S_2)}{S_2}\right)\right], \qquad S_i = \sqrt{\frac{2m}{\hbar^2}U_i}, \quad i = 1,2. \tag{26}$$

Square brackets indicate taking the integer part.

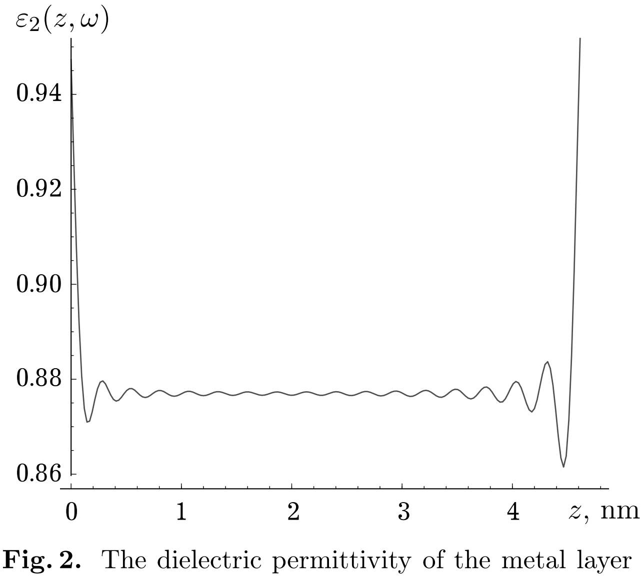

Fig. 2. The dielectric permittivity of the metal layer ($L = 5\,\mathrm{nm}$) when $\frac{\omega}{\omega_p} = 4$.

Expression for $\varepsilon(\boldsymbol{q}, z, z', \omega)$ (7) for the model (15) has the form (for details see [7])

$$\varepsilon_2(0,z,z',\omega) = \varepsilon_2(z,\omega)\,\delta(z-z') = \left(1 - \frac{\omega_p^2}{\pi n_e \omega^2}\sum_{n=1}^{n_{\max}}(k_F^2 - k_n^2)|\phi_n(z)|^2\right) \times \delta(z-z'). \tag{27}$$

The results of numerical calculations of $\varepsilon_2(z,\omega)$ for specific values $U_1 = 4.2\,\mathrm{eV}$ and $U_2 = 5\,\mathrm{eV}$, which correspond to the dielectrics (1 — polyethylene, 2 — SiO_2), are shown in Fig. 2.

The results obtained have shown that the dielectric function $\varepsilon_2(z,\omega)$ is different from constant only near the contact areas ($z = L$ and $z = 0$) (Fig. 2). This allows making some simplifications when studying the system (11)–(13).

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4. Investigation of the influence of the thickness of metal film on the wave spectrum

To solve the system of wave equations (11)–(13), Eq. (27) will be assumed that

$$\varepsilon_2(z,z',\omega)=\big(\varepsilon_2(L,\omega)+\alpha\Delta\varepsilon_2(z,\omega)\big)\delta(z-z'), \tag{28}$$

where

$$\varepsilon_2(L,\omega)=\frac{1}{L}\int_0^L\varepsilon_2(z,\omega)dz=1-\frac{\omega_p^2}{2\pi n_e\omega^2}\sum_{n=1}^{n_{\max}}(k_F^2-k_n^2)\overline{|\phi_n(z)|}^2, \tag{29}$$

$$\begin{aligned}\overline{|\phi_n(z)|}^2&=\frac{1}{L}\int_0^L|\phi_n(z)|^2dz\\&=|A|^2\bigg(\frac{1}{2}\Big(\frac{\chi_1^2}{k^2}+1\Big)+\frac{\chi_1}{2k^2L}\Big(1-\cos(2kL)\Big)+\frac{1}{4L}\Big(\frac{1}{k}-\frac{\chi_1^2}{k^3}\Big)\sin(2kL)\bigg).\end{aligned} \tag{30}$$

Substitution of (29) into (12) yields

$$\frac{\partial^2H_y(z,\omega)}{\partial z^2}+\big(k_0^2(\varepsilon_2(L,\omega)+\alpha\Delta\varepsilon_2(z,\omega))-k_x^2\big)H_y(z,\omega)=0. \tag{31}$$

The solution of the equation (31) for $H_y(z,\omega)$ can be found as expansion into a series in increasing powers of α

$$H_y(z,\omega)=\sum_{m=0}^{\infty}\alpha^mH_{y,m}(z,\omega). \tag{32}$$

In particular, the first two equations $H_{y,0}(z,\omega)$ and $H_{y,1}(z,\omega)$ are following:

$$\frac{\partial^2H_{y,0}(z,\omega)}{\partial z^2}+\big(k_0^2\varepsilon(L,\omega)-k_x^2\big)H_{y,0}(z,\omega)=0, \tag{33}$$

$$\frac{\partial^2H_{y,1}(z,\omega)}{\partial z^2}+\big(k_0^2\varepsilon(L,\omega)-k_x^2\big)H_{y,1}(z,\omega)=-k_0^2\alpha\Delta\varepsilon_2(z,\omega)H_{y,0}(z,\omega). \tag{34}$$

When modeling the influence of the thickness of a metal film L on SPPs spectrum, we will limit ourselves to the case $H_y(z,\omega)\approx H_{y,0}(z,\omega)$. In this case, the dispersion relation has the form

$$e^{-4k_1\frac{L}{2}}=\frac{k_1/\varepsilon_1+k_2/\varepsilon_2}{k_1/\varepsilon_1+k_2/\varepsilon_2}\frac{k_3/\varepsilon_1+k_3/\varepsilon_2}{k_3/\varepsilon_3+k_2/\varepsilon_2}, \tag{35}$$

$$k_i^2=k_x^2-k_0^2\varepsilon_i,\ \ i=1,2,3;\ \ k_0=\frac{\omega}{c}, \tag{36}$$

which coincides with the results obtained in Ref. [1]. Here $\varepsilon_1=\varepsilon(\omega)$, $\varepsilon_2=\varepsilon(L,\omega)$, and $\varepsilon_3=\varepsilon(\omega)$.

Similarly as in our previous work [11], for the upper layer we took a polyethylene with a permittivity constant $\varepsilon_1=2.3$ and the electron work function $U_1=4.24\,\mathrm{eV}$; SiO_2 for the lower dielectric substrate with a permittivity constant $\varepsilon_1=4$ and the electron work function $U_2=5\,\mathrm{eV}$. The dielectric function of the metal layer (gold) is described by the function (16)

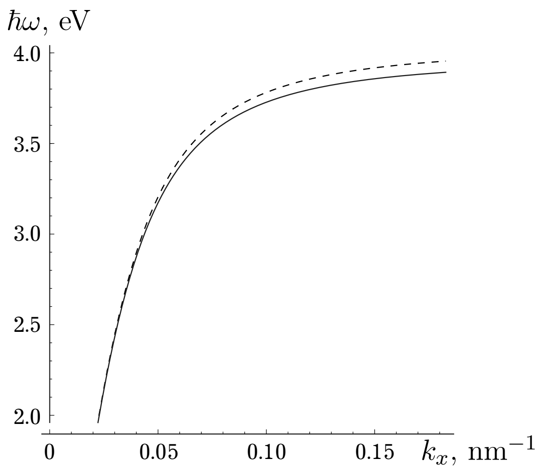

Fig. 3. SPPs spectrum for the Drude model $\varepsilon^D(\omega)$ (dotted line) and $\varepsilon_2(L,\omega)$ (solid line).

$$\varepsilon_2(L,\omega)=1-\frac{\omega_p^2}{2\pi n_e\omega^2}\sum_{n=1}^{n_{\max}}(k_F^2-k_n^2)\overline{|\phi_n(z)|}^2. \tag{37}$$

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Fig. 3 shows the result of spectrum calculations obtained from (35) using (29), (30). These results we compared to the data obtained for the Drude model with negligible damping [1]

$$\varepsilon(L,\omega)=\varepsilon^{D}(\omega)=1-\frac{\omega_p^2}{\omega} \tag{38}$$

in which the plasmon frequency spectrum does not depend on the thickness L.

In Table 1 the results of numerical calculations of the dependence of frequency $\omega^*=\frac{\omega}{\omega_p}$, and a wave vector $k_x^*=\frac{k_x c}{\omega_p}$ on the number of levels of dimensional quantization $n_{\max}$ are provided [12].

Table 1.

	$\varepsilon_2(L,\omega^*)$	$\varepsilon^D(\omega^*)$	
L (nm)	ω^*	ω^*	$n_{\max}$
$k_x^*=1$			
100	0.33992328	0.34237082	335
1000	0.34016832	0.34237082	3357
5000	0.34019295	0.34237082	16789
10000	0.34019603	0.34237082	33579
30000	0.34019749	0.34237082	100737
$k_x^*=2$			
100	0.40974355	0.41421356	335
1000	0.41021317	0.41421356	3357
5000	0.41025793	0.41421356	16789
10000	0.41026353	0.41421356	33579
30000	0.41026618	0.41421356	100737
$k_x^*=3$			
100	0.42679347	0.43187178	335
1000	0.42732626	0.43187178	3357
5000	0.42737706	0.43187178	16789
10000	0.42738341	0.43187178	33579
30000	0.42738642	0.43187178	100737
$k_x^*=4$			
100	0.43313236	0.43844718	335
1000	0.43368968	0.43844718	3357
5000	0.43374282	0.43844718	16789
10000	0.43374947	0.43844718	33579
30000	0.43375261	0.43844718	100737

The data in the table show that difference between spectra $\omega(k_x)$ for both models increases with increasing of the wave vector k_x. Also, it should be noted that with increasing of L, $\omega(k_x)$ is steadily approaching from below the values obtained for the Drude model.

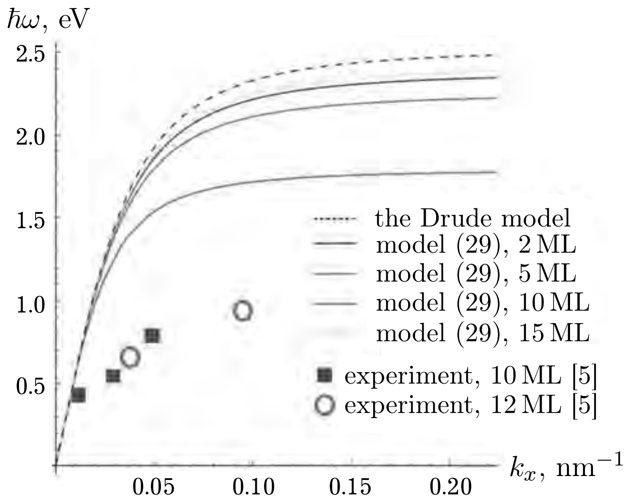

Fig. 4. SPPs spectrum for the Drude model $\varepsilon^D(\omega)$ (dotted line) and $\varepsilon_2(L,\omega)$ for the different thicknesses of a metal layer (solid lines), 1 ML $\sim$ 0.24 nm.

In Fig. 4, the results of calculations for the structure "Si–silver–SiO_2" are shown. The dielectrics were simulated for the following parameters: $U_1=5\,$eV, $\varepsilon_1=2.4$ and $U_1=4.8\,$eV, $\varepsilon_1=11.7$ for Si and SiO_2 correspondingly [1].

The same figure shows the experimental results for the structure "Si–silver–SiO_2" published in [5]. These results demonstrate that the spectrum of plasmons strongly depends on the thickness of a metal layer when $L\sim 50\,$ML. As can be seen from Fig. 4, the proposed approach gives a qualitative explanation of the influence of a metal film on the SPPs spectrum.

Note that the results obtained in [5] for the SPPs spectrum were obtained for structures that consist of 2–15 monolayers, thus simulation of $\varepsilon_2(\boldsymbol{q},z,z',\omega)$ should be carried out for such thicknesses. In such metal structures, quantum effects become significant [9,10,13] and they should be taken into account.

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Поверхневі плазмон–поляритони в структурах "діелектрик–метал–діелектрик": вплив товщини металевого прошарку

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Запропоновано та досліджено модель, яка дає змогу пояснити експериментальні дані щодо впливу товщини металевого прошарку на спектр SPP хвиль у гетерогенних структурах "діелектрик–метал–діелектрик".

Ключові слова: *поверхневі плазмони, спектр плазмона, товщина металевого шару, діелектрична проникність.*

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